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Ferroelectric Electron Emission: Principles and Technology

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Abstract

The spontaneous electrical polarization of ferroelectric materials can be changed either by reversal or by phase transition from a ferroelectric into a non-ferroelectric state or vice versa. If spontaneous polarization changes are induced with fast heat, mechanical pressure, laser or electric field pulses on a submicrosecond time scale, strong uncompensated surface charge densities and related polarization fields are generated, which may lead to the intense self-emission of electrons from the negatively charged free surface areas of the ferroelectric sample. Hence, electron guns can be built with extraction-field-free ferroelectric cathodes, which may be easily separated from the high-field regions of post-accelerating gap structures. The intensity, the energy, the temporal and spatial distribution, and the repetitition rate of the emitted electron beams can be controlled within wide limits via the excitation pulses and external focusing and accelerating electromagnetic fields. The technological advantages and difficulties of ferroelectric cathodes during production and in practical operation are identified and discussed. A new design of electron guns with ferroelectric cathodes is described. Experience with a few applications of ferroelectric electron emission is reported and suggestions for further applications are made.

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1. Introduction

In 1984 Rosenman et al. [1] reported for the first time on the experimental observation of weak electron emission (10^{-12} A/cm^2) during polarization reversal of ferroelectric (FE) material. Strong electron emission up to 100 A/cm^2 from ferroelectric material was detected at CERN in 1988 and has since been discussed and used in several applications [2]–[9]. Confusion has arisen in defining what is meant by 'ferroelectric electron emission', owing to work with different methods of electron emission from FE in other laboratories [10]–[13]. The author feels obliged to clarify the term 'FE emission', which is discussed in this paper and which corresponds to the 'strong FE emission' referred to above. In Refs. [2]–[8], dealing mostly with the principles and features of this typical FE emission, the observations made experimentally and the performance limits in some applications have been discussed. This paper will also deal with some of the technological requirements and difficulties arising when designing and manufacturing FE cathodes for a particular purpose. The intrinsic features of FE emission differ significantly from classical electron emission mechanisms, which all require external potentials to extract electrons from the cathode. FE emission does not need extraction fields and, hence, allows the design of electron gun structures, which cannot be carried out with classical electron sources.

2. Basic features of FE emission

FE emission resembles friction-induced emission from insulators and is based on the macroscopic separation of charges of opposite polarity onto opposite surfaces of an FE sample (Fig. 1). The charge separation can be induced by applying a fast temperature rise [2], a mechanical pressure pulse (formerly used in fire lighters), a laser pulse [5] or a high-voltage (HV) pulse [3], [4], and [6] to the FE sample. With all these different excitation methods a fast change (switching) of the spontaneous FE polarization P_S is induced, which is equivalent to the momentary appearance of high surface charge densities. The surface charge densities may be so great, that, as a result of the strong self-field, electrons are spontaneously emitted from the negatively charged FE surface as shown in Fig. 1c. In case of electric field excitation, an HV pulse of negative polarity with respect to the emitting surface must consequently be applied to the opposite surface of the FE cathode. The emitting surface is not exposed to an external extraction field before polarization switching, hence, the resulting ejection of electrons may be considered as a 'self-emission' process. The electrons are emitted from the FE surface into the adjacent vacuum or gas-filled volume with finite kinetic energies, which may reach values above 100 keV [1], [2], [8], and [14]. It is important for technical applications that FE emission, other than friction-induced emission from insulators, can be easily controlled in space and time [4]. Since the rapid change of spontaneous polarization P_S is characterized by a typical hysteresis curve, the FE electron emission can only start above a defined threshold of thermal, mechanical, optical, or electrical excitation energy. FE emission is only useful when the excitation thresholds are lower than the destruction and breakdown limits of the FE material and of the metallic surface electrodes used. The density distribution of switchable FE domains determines the average strength of electron emission. The spontaneous polarization of an

individual domain in mono- and polycrystalline FE material is generally fully and almost instantaneously switched from one direction to another. Hence, even at weak average emission current, high kinetic electron energies are observed [1], [2], and [5]. Therefore it is physically meaningless to distinguish between 'low and high emission current regimes'. Strong emission is achieved when a large number of individual domains are switched together within a short (subnanosecond) time interval. Domain size may vary between millimetres in single crystals and nanometres in polycrystalline FE material. The emission process, when induced by HV pulses, is strongly dependent on the electrode arrangement and on the electric field distribution inside the FE sample before [15] and after switching. The electrically induced emission from FE samples covered with metallic grid electrodes is very efficient, but emission is also obtained from FE surfaces without any electrode or conducting material [16]. As in the case of any other type of electron emission, FE emission mechanisms also provoke the formation of surface plasma if the local emission strength becomes so great that material is released from the emitting surface. The plasma is generated by electron sputtering, hopping, and by excitation and ionization of the surface atoms and molecules under the influence of the local surface fields, e.g. in the vicinity of the edges of a metallic grid electrode. Following the typical field configuration on the emitting surface, the plasma spreads rapidly across the cathode, while acting as a transparent electrode for the emitted electrons and enhancing FE switching and emission. After switching, on the other hand, the polarization field on the bare FE surface holds back the plasma ions from escaping with the departing electrons and serves as a reservoir for restoring compensating charges to the emitting surface. Care must be taken after surface plasma formation not to load the sample with too much excitation energy in excess of the necessary energy for polarization switching. Otherwise the risk of destroying the grid electrodes and the FE sample surface increases. Similarly, strong d.c. or low-frequency external (extraction) fields should not be used together with strong FE emission, since the plasma formation will be enhanced and its expansion into the direction of the emitted electron beam will lead to breakdown in the beam transport structure. The use of HV pulses with negative polarity to the grid electrode on the emitting cathode surface, or with positive polarity to the opposite side, is generally destructive for most applications. In certain polycrystalline FE materials, such as zirconium-rich PLZT (lead-lanthanum-zirconium-titanate) ceramics, FE microdomains can be formed or annihilated at a subnanosecond time scale under the influence of local electric fields in a rather large temperature range above and below the FE/non-FE phase transition temperatures [2]. Very fast switching response times down to 0.1 ns were observed in laser-induced self-emission experiments from FE photocathodes [8]. Figure 2a shows the emission pattern for a weakly emitting PLZT 2/95/5 sample characterized by a double peak emission due to forward switching and microdomain formation (first peak) and to depolarization and microdomain annihilation or compensation (second peak), when the excitation field decays. In a strongly emitting sample both peaks may merge into a single emission pulse (Fig. 2b). In both cases the second electron pulse (by depolarization) is not caused by emission from the bare FE surface, but from the grid electrode, enhanced by the surface plasma, and accelerated by the surface field remaining from the HV excitation pulse to the rear electrode.

3. Technological aspects

Though the basic scheme of FE electron emission appears simple and though strong emission has been observed with many FE materials showing strongly differing phase diagrams, the success or failure of achieving strong FE emission as defined above depends on many parameters and environmental factors. Only a few of them could be examined more carefully and are dealt with below. The choice of the FE cathode material, the preparation methods, and the geometry of samples and excitation electrodes described in earlier references [3], [4], and [7], though not optimized, favoured strong FE emission. The density of switchable FE domains is very sensitive to heat treatments during the material production and sample preparation processes. Also the grain size and the corresponding domain size (generally two orders of magnitude smaller) are determined by the heat treatments during production and preparation of an FE cathode. In polycrystalline PLZTs a grain size between 1 and 10 μm appeared as a reasonable choice. The generation of high oxygen vacancy concentrations at high temperatures may raise switching and emission thresholds so far that the excitation of emission becomes difficult. Oxygen vacancies and other crystalline defects and impurities can act as internal space-charge centres, which also interfere with the charge compensation processes before and after strong polarization changes. The size and geometry of the grid (GE) and rear (RE) electrodes on the FE cathode together with its thickness (Fig. 3), determine the field distribution on the emitting surface before and after polarization switching [15]. Therefore the electrodes must be matched to thick [3] and thin-film FE samples differently. Important parameters are FE porosity and surface roughness: if the surface is too smooth the electrodes adhere badly, if it is not well polished or etched, the field enhancement at holes or scratches may lead to sample perforation by the applied HV pulses. Significant influence is also exerted by the applied electroding procedures (resulting from different heat treatments, electrode materials, micromechanical electrode structure), for example, metal evaporation, sputtering, screen printing, or galvanization. All procedures may work provided the FE material is not destroyed or strongly modified, e.g. by overheating and diffusion processes. The geometry and the pressure of the HV-pulse contacts to the FE electrodes may strongly influence the switching thresholds under the pressurized regions under the contacts and the field distribution near the emitting grid after polarization switching. The thresholds for switching and electron emission can be lowered by careful heat treatments, prepoling at temperatures just above the ferroelectric-paraelectric phase transition temperature and by weak surface metallization with thin, non-contiguous layers. Though FE electron emission was observed in vacuum, in low-pressure gas, in dense plasma and even in oil, the environmental effects have to be carefully taken into account. The main requirements of FE cathodes for an application are:

a) control of emission precisely in time and space, control of repetition rate, reproducibility, reliability and fatigue resistance, robustness under severe environmental conditions in a large temperature range;

b) optimization of maximum beam current amplitude or total emitted charge for a given application;

c) electron beam quality: control of homogeneity, energy spread, brightness, focusing properties of the electron beam pulses.

The control is exerted by the cathode design and preparation including the well-matched choice of FE material. The electrical circuits for FE excitation and electron beam acceleration must be adapted to the enviroment and the sample parameters to guarantee a sub-microsecond HV rise time on the cathode and to avoid breakdowns in the electron beam transport structure. In the past, mostly solid disks of a few tenths up to 1 mm thickness have been used by the author as FE cathodes. In Ref. [17] cathodes have been prepared, which consist of a mixture of FE powder with a non-FE binding material. These simple and cheap cathodes, which resemble to some extent the zirconium-rich PLZT cathodes containing FE domains in a more-or-less non-FE matrix, have been operated successfully (see Fig. 4).
Characteristics of laser-induced emission have been studied and reported in Refs. [5], [6], [8], and [18]. A large part of the results covers experiments under various laser pulse conditions with FE photocathodes used in the classical way: laser-illumination under d.c. extraction field. It appears that this scheme of employing FE photocathodes is the least interesting one. The use of enhanced and self-emission methods opens the possibility of operating FE photocathodes at IR wavelength and at ultra-short laser pulse lengths with high efficiency and unequalled reliability.

4. Applications and technical problems

The first application of strongly emitting FE cathodes was their use as trigger elements to ignite the discharge of high-power gas switches described in Refs. [19], [6], and [7]. Emitted beam quality and intensity were less important than high precision and robustnes in an environment characterized by high power dissipation, metal plasma formation, and metal deposition. A more demanding application with respect to precision, repetition rate, and beam quality is the alternating gradient space-charge focusing of low-energy, heavy-ion beams described in Ref. [9] with pulsed electron beams generated with an electron gun equipped with an FE cathode (Fig. 5). In contrast with conventional electron guns, this structure features an extraction-field-free beam expansion volume adjacent to the FE cathode surface. Electrostatic and magnetic focusing can be applied before the beam reaches the post-accelerating gap. Figures 6a and 6b show the focusing and high repetition rate capabilities of this type of gun [4], [9]. Also laser-driven electron guns with FE cathodes can be used following the same principle of Fig. 5. The FE photocathodes have, however, to be excited in self-emission mode and extraction fields must be pulsed or alternating with very high frequency if short pulses are desired and if vacuum breakdowns are to be avoided. PLZT photocathodes have been illuminated with femtosecond laser pulses of 0.8 mJ energy and 2 TW/cm^2 peak power density at a repetition rate of 1.5 kHz. Thin-layer FE emission was proposed in 1989 [20] and development work has been described in Refs. [11], [14] and [15]. FE self-emission functions with very low excitation voltage amplitudes (< 100 V), but only above a minimum FE layer thickness, which is determined by the mutual attraction of the separated charge layers formed on opposite sides of the cathode. For thinner layers extraction potentials must be applied [11], [14]. Many techniques useful for the production and preparation of thin-layer FE cathodes are available from electronic thin-film and FE memory technologies. Simple and cheap FE powder cathodes [17] are especially suited for large-area emission applications, e.g. for large volume

ionization processes such as exhaust gas transmutation. They may also be employed in electron guns with a bent emitting cathode surface for any purpose. Shaped (for example conical) cathodes of this type may be used to focus electron beams just by cathode geometry without electric or magnetic focusing fields.

5. Conclusion

A large range of applications can be identified covering machining applications, electron guns for low- and high-energy accelerator technology, thin-layer applications (ultra-flat TV screens microlasers), photocathodes at IR wavelengths, power tubes for RF, microwave and X-ray generation, and large volume ionization. More R&D is required for specific applications to optimize performance and to solve the technological and material problems, which have been briefly mentioned in this paper. However, the starting conditions are promising, since many technological procedures can be taken over from electronics, from classical ferroelectrics, from thin-film technology and from high-T_c superconductor manufacturing.

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Figure captions

Fig. 1: Conventional emission (a) and FE emission by polarization switching and macroscopic charge separation (b,c).

Fig. 2: Characteristic electron pulse currents from weakly (a) (time base 125 ns/div) and strongly (b) (500 ns/div) emitting FE cathodes.

Fig. 3: Typical excitation electrode geometries for axial (a) and hollow beams (b).

Fig. 4: Tubular FE-powder cathode system for electron irradiation of large gas volumes (GE = grid, RE = rear, and EX = extraction electrodes).

Fig. 5: Typical FE electron gun design with extraction-field-free cathode surface.

Fig. 6: Focusing (a) and repetition rate (b) characteristics of the FE electron gun.

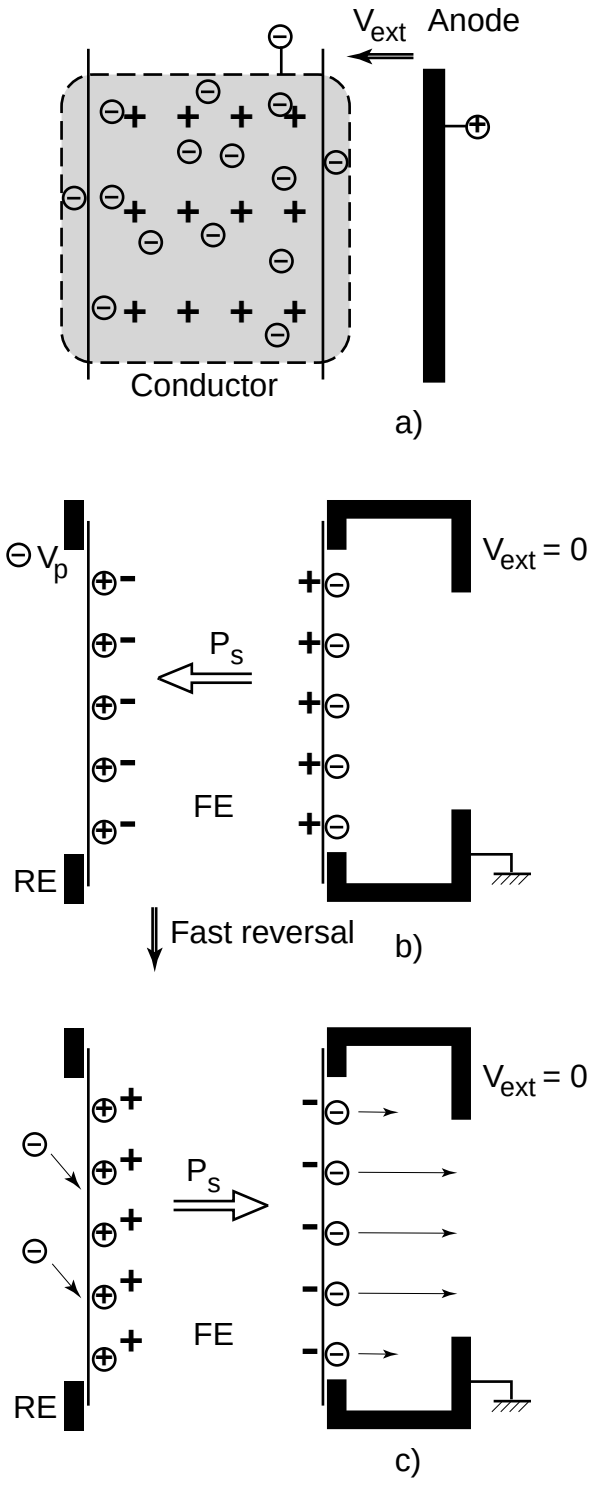

V_ext
Anode
Conductor
a)
V_p
P_s
FE
RE
V_ext = 0
Fast reversal
b)
P_s
FE
RE
V_ext = 0
c)

Figure 1

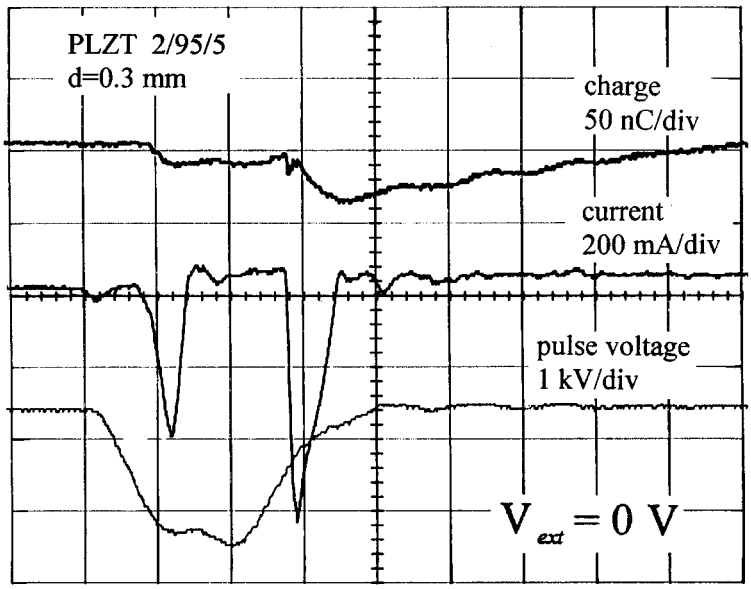


Figure 2(a)

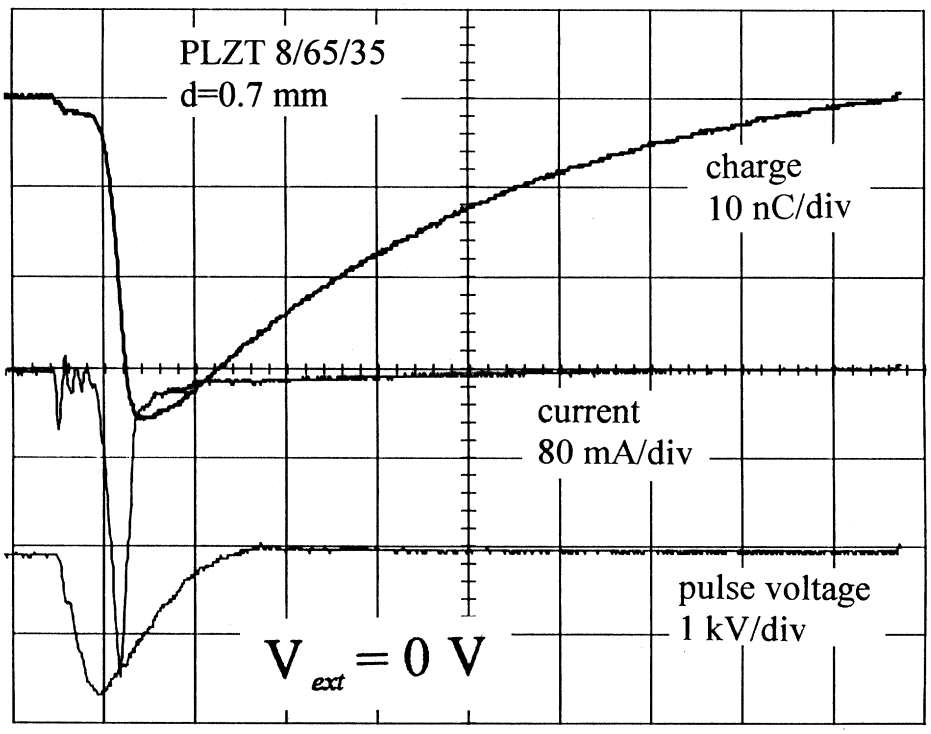


Figure 2(b)

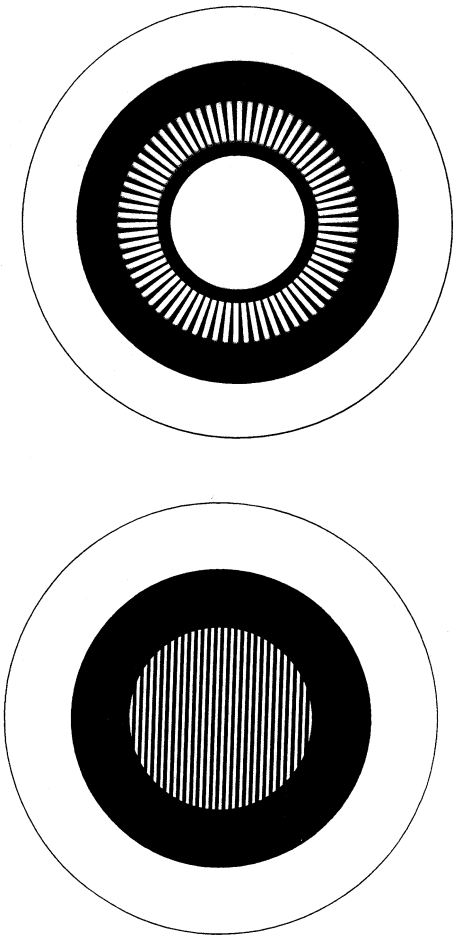


Figure 3

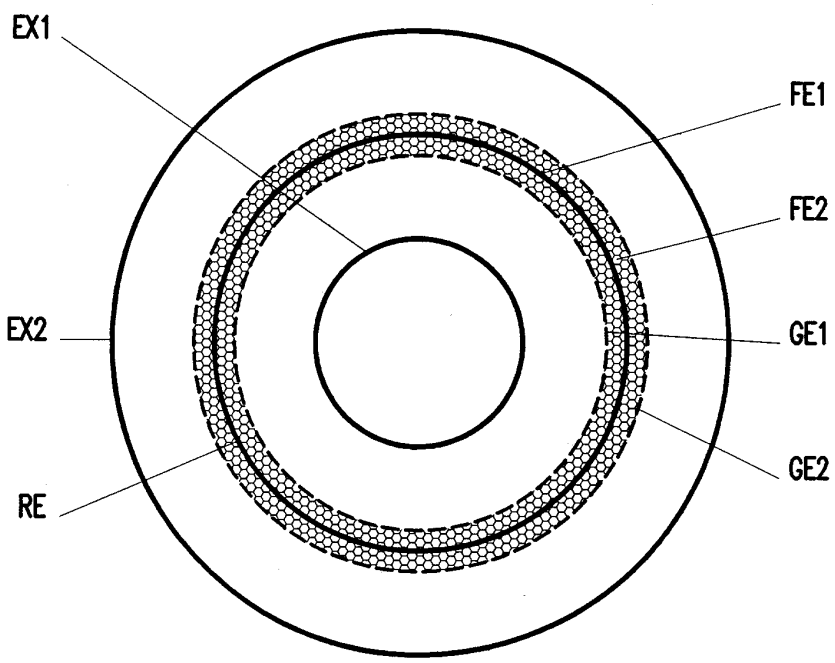


Figure 4

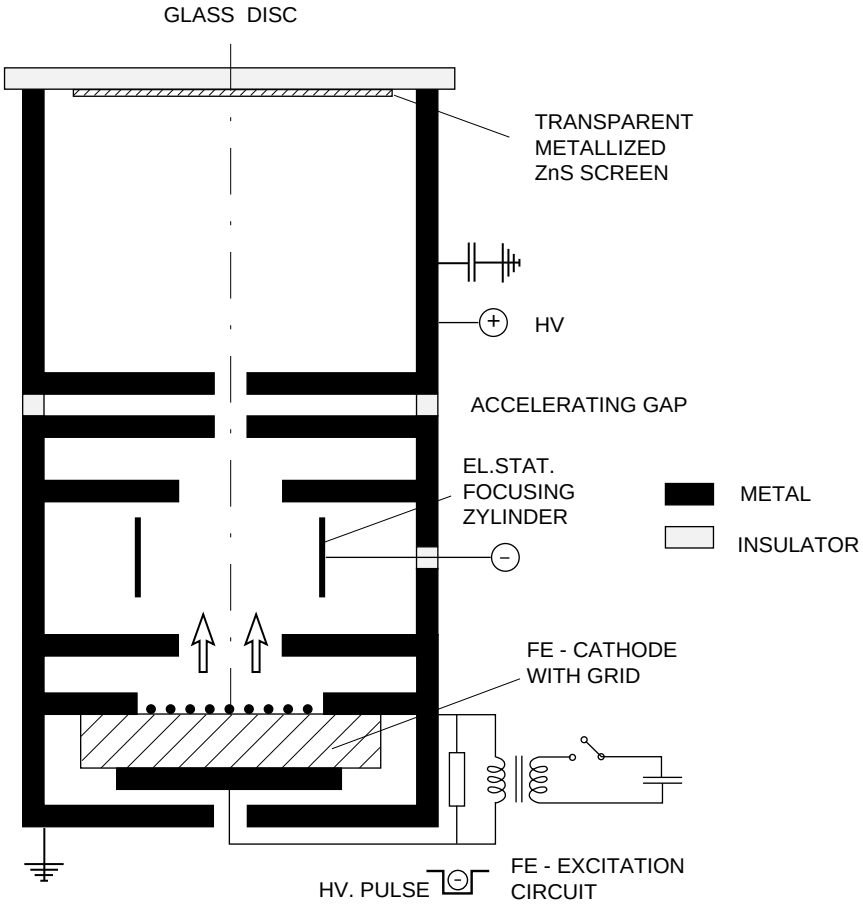
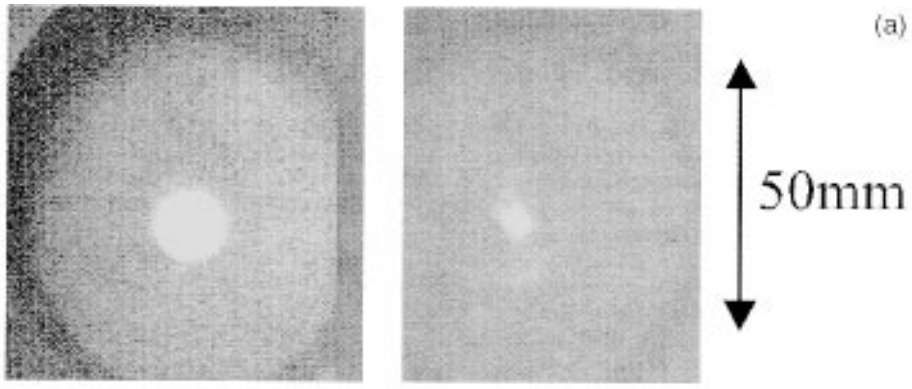
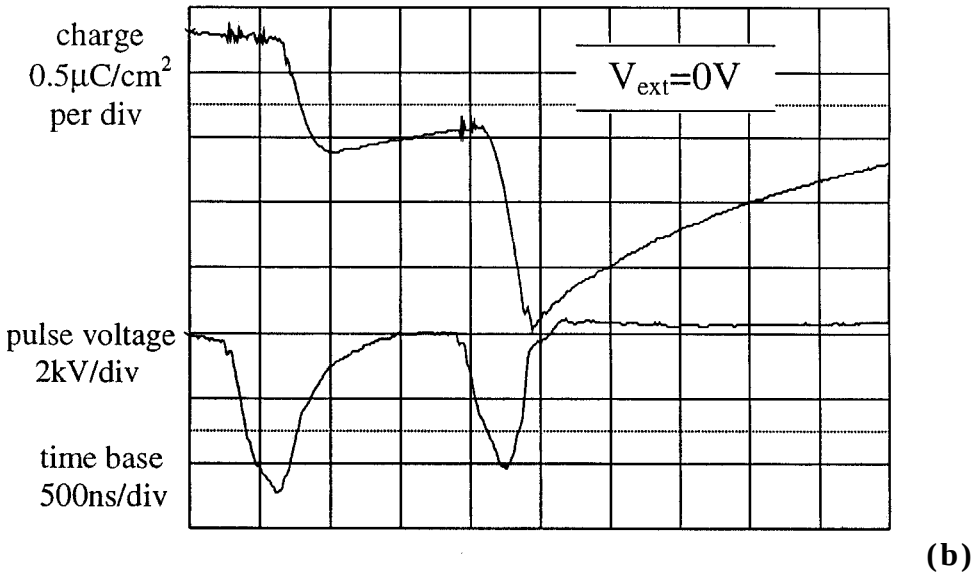

GLASS DISC
TRANSPARENT
METALLIZED
ZnS SCREEN
HV
ACCELERATING GAP
EL.STAT.
FOCUSING
ZYLINDER
METAL
INSULATOR
FE - CATHODE
WITH GRID
HV. PULSE
FE - EXCITATION
CIRCUIT

Figure 5


(a)
50mm


charge
0.5μC/cm²
per div
V ext=0V
pulse voltage
2kV/div
time base
500ns/div

(b)

Figure 6